

Spin-1/2 formalism for two-level problems

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The formalism of the theory of angular momentum (spin-1/2) is the most appropriate for the treatment of the two-level problem (interaction of radiation with a quantum mechanical system with an isolated pair of eigen states of energy). The density matrix and the total Hamiltonian can be expressed as vectors in a co-ordinate system spanned by the basic spin representations related to spin space. The dynamical problem is solved by finding from the equation of motion the spinor transformation T which effects the rotation of the density operator.

The spin-1/2 formalism shows the interconnections among the distinctive methods of treating the problem (due to Feynman & Vernon 1957, Dicke 1954, and Lamb 1964). An *analytic* solution of the mechanical problem of finding the polarization for the transition $\Delta m = 0$ is presented. Expressions are given for electrical field, power, and oscillation frequency in terms of the one variable, the pulse strength θ_N determined by the density N of molecules interacting with the radiation field in a stationary process. The matrix of interaction containing this parameter is a rotation about the 1-axis in spin-space through the pulse angle θ and operates on the initial system vector to produce the system vector at time t . The latter is a matrix whose elements are the expectation values of the unperturbed Hamiltonian and polarization.

INTRODUCTION

The two-level problem dealing with spontaneous and induced emission of radiation from an assembly of atoms (or molecules) with an isolated pair of low-lying energy levels has been treated in several ways in order to obtain the emitted power and frequency shifts. Of the four general forms of the equation of motion in quantum mechanics, with which the two-level problem can be treated, namely, those involving the Schrödinger wave function ψ , the density operator ρ , the T -operator, and the Heisenberg variables, only the Heisenberg form seems to have been left out. Some of these methods have been extended and refined to bring out the more detailed, and non-linear aspects of the process (Mohanty 1967, Roy 1969).

There is a very simple form of the dynamical law brought out by Feynman & Vernon (1957) specifically for the two-level problem. This presents the dynamical problem as the motion of a gyroscope (a vector \vec{r}) with frequency ω in r -space. Dicke (1954) presents no specific form of the equation of motion for the two-level problem. He proceeds on intuitive grounds to deal with an important aspect of the radiative process, namely, the existence of certain exceptional states of the gaseous assembly of N molecules and shows that in a spontaneous emission

process the intensity of radiation is proportional to N^2 (and not N as for independent molecules).

By a juxtaposition of Pauli operators with Feynman frequencies and vector $\rightarrow r$, one can express the Hamiltonian and density operators respectively as vectors in spin space. One can almost characterize the two-level problem as a spin-1/2 problem. Hints on the application of spin-1/2 formalism to two-level problems have appeared in literature at various times but the authors feel that there is no systematic treatment of the problem from this angle. The purpose of this paper is to present such a treatment and to derive Feynman's solutions *analytically*. Further, the main postulates of Dicke are shown to follow from a general form of the dynamical law. In this way the interconnections between the methods of Lamb (1964), Feynman & Vernon (1957) and Dicke (1954) can be shown.

The system considered is a beam of molecules with an isolated pair of energy levels entering a cavity tuned to the molecular transition frequency, the molecules being all in the state of higher energy just when they enter the cavity at $t = 0$. The proper values of the energy are E_a and E_b , the molecular transition frequency being $\omega_0 = \hbar^{-1}(E_a - E_b)$.

SPIN SPACE

Let ψ represent an arbitrary state of the system

$$\psi = a\psi_a + b\psi_b \quad (1)$$

where the co-efficients satisfy the normalization condition

$$aa^* + bb^* = 1 \quad (2)$$

The matrix

$$T = \begin{pmatrix} a & -b^* \\ b & a^* \end{pmatrix} = \begin{pmatrix} \alpha & \beta \\ \gamma & \delta \end{pmatrix} \quad (3)$$

and its conjugate imaginary

$$\bar{T} = \begin{pmatrix} a^* & b^* \\ -b & a \end{pmatrix} \quad (3a)$$

satisfy $TT = 1$ in virtue of the normalization (2). Hence T is unitary. (1) represents the unitary transformation of ψ_a by T :

$$\begin{pmatrix} a & -b^* \\ b & a^* \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} a \\ b \end{pmatrix}$$

The elements of the matrix T are just the Cayley-Klein parameters.

In general the matrix T transforms a two-component wave function $\begin{pmatrix} A \\ B \end{pmatrix}$ into another $\begin{pmatrix} A' \\ B' \end{pmatrix}$ as follows :

$$\left. \begin{aligned} A' &= aA - b^*B = \alpha A + \beta B \\ B' &= bA + a^*B = \gamma A + \delta B \end{aligned} \right\} \quad \dots \quad (4)$$

subject to the condition (2), i.e.,

$$\alpha\delta - \beta\gamma = 1 \quad \dots \quad (2a)$$

A spinor is a two-component quantity defined by the linear transformation (4) subject to (2) (or (2a)) when the co-ordinate system is rotated. The problem of finding ψ is equivalent to finding a representative of T involving the interaction which effects a rotation in spin space.

In Euler resolution the general rotation T is given by the product

$$T = T_\psi T_\theta T_\phi$$

where T_ψ , T_θ , T_ϕ are unitary matrices effecting rotations in spin space through the Euler angles, ϕ , θ , ψ and are given by

$$\begin{aligned} T_\phi &= I \cdot \cos \phi/2 + i\sigma_3 \sin \phi/2 = e^{i\phi/2\sigma_3} \\ T_\theta &= I \cos \theta/2 + i\sigma_1 \sin \theta/2 = e^{i\theta/2\sigma_1} \\ T_\psi &= I \cdot \cos \psi/2 + i\sigma_3 \sin \psi/2 = e^{i\psi/2\sigma_3} \end{aligned} \quad \dots \quad (5)$$

The Pauli operators $\vec{\sigma} = (\sigma_1, \sigma_2, \sigma_3)$ obey the commutation rules

$$\sigma_1\sigma_2 + \sigma_2\sigma_1 = 0; \quad \sigma_1\sigma_2 - \sigma_2\sigma_1 = 2i\sigma_3 \text{ etc.}$$

Also

$$\sigma_1\sigma_2 = i\sigma_3 \text{ etc.}, \quad \sigma_1^2 = \sigma_2^2 = \sigma_3^2 = I$$

They are represented by $\sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$, $\sigma_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$, $\sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$

Let us denote the unit matrix by $\sigma_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$

THE DENSITY MATRIX

The density operator is given by $\rho(t) = \begin{pmatrix} a^* & a & b^* & a \\ a^* & b & b^* & b \end{pmatrix} \quad \dots \quad (6)$

Introducing a new operator $\mu(t) = 2\rho(t) - 1$ so that $\mu(0) = \sigma_3 \quad \dots \quad (7)$

at $t = 0$, the dynamical problem reduces to finding the transformed operator

$$\bar{T}T\sigma_3 = \sigma_n = r_1\sigma_1 + r_2\sigma_2 + r_3\sigma_3 \quad \dots \quad (8)$$

where σ_n is the component of σ in a general direction n with direction cosines r_1, r_2, r_3 . Since

$$1 = T\sigma_3^2\bar{T} = T\sigma_3\bar{T}T\sigma_3\bar{T} = \sigma_n^2$$

r_1, r_2, r_3 are also the components of a vector \vec{r} of unit length

$$r_1^2 + r_2^2 + r_3^2 = 1 \quad \dots (9)$$

Using (3) and (3a) we obtain r from the transformation (8)

$$\begin{aligned} r_1 &= ab^* + a^*b, \\ r_2 &= i(ab^* - a^*b), \\ r_3 &= aa^* - bb^* \\ r_0 &= 1 = aa^* + bb^* \end{aligned} \quad \dots (10)$$

(9) follows from (10) when the normalization condition is used. The density matrix may then be written as

$$\rho = \frac{1}{2} \begin{pmatrix} 1+r_3 & r_1-ir_2 \\ r_1+ir_2 & 1-r_3 \end{pmatrix} = \frac{1}{2}\sigma_0 + \frac{1}{2}(r_1\sigma_1 + r_2\sigma_2 + r_3\sigma_3) \quad \dots (11)$$

and

$$\mu = \begin{pmatrix} r_3 & r_1-ir_2 \\ r_1+ir_2 & -r_3 \end{pmatrix} = r_1\sigma_1 + r_2\sigma_2 + r_3\sigma_3 \quad \dots (12)$$

In term of the spin angular momentum $\vec{S} = \frac{1}{2}\hbar\vec{\sigma}$, $S_0 = \frac{1}{2}\hbar\sigma_0$

$$\rho = \hbar^{-1}(r_1S_1 + r_2S_2 + r_3S_3 + r_0S_0)$$

The form of the matrix μ shows that μ is a vector of unit length in the space spanned by the unit vectors $\sigma_1, \sigma_2, \sigma_3$ and rotating under the influence of the radiation field.

THE HAMILTONIAN

The unperturbed Hamiltonian is represented by the matrix

$$H_0 = \begin{pmatrix} E_a & 0 \\ 0 & E_b \end{pmatrix}$$

If the energies are measured from their mean taken as zero, then

$$H_0 = \frac{1}{2}\hbar\omega_0 \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} = \frac{1}{2}\hbar\omega_0\sigma_3$$

If the system has no permanent electric dipole moment the perturbation is represented by the matrix

$$V = \begin{pmatrix} 0 & V_{ab} \\ V_{ba} & 0 \end{pmatrix}$$

Introducing the real quantities

$$\begin{aligned} \omega_1 &= \hbar^{-1}(V_{ab} + V_{ba}) \\ \omega_2 &= i\hbar^{-1}(V_{ab} - V_{ba}) \\ \omega_3 &= \omega_0 \end{aligned} \quad \dots (13)$$

We have

$$V = \frac{1}{2}\hbar \begin{pmatrix} 0 & \omega_1 - i\omega_2 \\ \omega_1 + i\omega_2 & 0 \end{pmatrix} = \frac{1}{2}\hbar(\omega_1\sigma_1 + \omega_2\sigma_2) \quad (14)$$

The total Hamiltonian is given by :

$$\begin{aligned} H &= H_0 + V = \frac{1}{2}\hbar(\omega_1\sigma_1 + \omega_2\sigma_2 + \omega_3\sigma_3) \\ &= \frac{1}{2}\hbar \begin{pmatrix} \omega_3 & \omega_1 - i\omega_2 \\ \omega_1 + i\omega_2 & -\omega_3 \end{pmatrix} \end{aligned} \quad (15)$$

THE EQUATION OF MOTION

The time development of ρ is given by

$$i\hbar \frac{\partial \rho}{\partial t} = -(\rho H - H\rho) \quad (16)$$

The commutation rules for $\sigma_1, \sigma_2, \sigma_3$ lead to the following relation for any two vectors \vec{a} and \vec{b}

$$(\vec{a} \cdot \vec{\sigma})(\vec{b} \cdot \vec{\sigma}) = \vec{a} \cdot \vec{b} + i(\vec{a} \times \vec{b}) \cdot \vec{\sigma} \quad \dots \quad (17)$$

which gives

$$(\vec{a} \cdot \vec{\sigma})(\vec{b} \cdot \vec{\sigma}) - (\vec{b} \cdot \vec{\sigma})(\vec{a} \cdot \vec{\sigma}) = 2i(\vec{a} \times \vec{b}) \cdot \vec{\sigma} \quad \dots \quad (17a)$$

Using this relation and substituting for ρ and H from (11) and (15) in (16)

$$i\hbar \frac{\partial}{\partial t}(\vec{r} \cdot \vec{\sigma}) = i\hbar(\vec{\omega} \times \vec{r}) \cdot \vec{\sigma}$$

or

$$\frac{\partial \vec{r}}{\partial t} = \vec{\omega} \times \vec{r} \quad \dots \quad (18)$$

This is *Feynman's equation* for the two-level problem. The equation is true for all co-ordinate systems (in spin space) obtained from each other by rotational displacements and for rotating co-ordinate systems.

The interaction process may be regarded as the motion of a vector of unit length in spin space and lends itself to a graphical solution considered by Feynman. The essential step is to transform to some rotating co-ordinate system where the vector \vec{r} appears to execute a simple precessional motion about the 3-axis, the component of the frequency $\vec{\omega}$ along the 3-axis being constant and the other two components being zero. The representation of the dynamical law as a unitary transformation (rotation) in spin space automatically leads to the graphical method although we shall adopt here an analytical procedure which is entirely parallel to the graphical method.

ROTATIONS

Let us denote the four co-ordinate systems here by Σ_S (space-fixed), Σ_B (body-fixed), Σ_R (rotating) and Σ_P (precessional). We pass over to Σ_R by means of T_ϕ effecting a rotation about the 3-axis of Σ_S through an angle $\phi(t)$. One can follow the motion of \vec{r} in Σ_S or regard \vec{r} as fixed in Σ_S and follow the motion of a co-ordinate system Σ_B relative to Σ_S . Alternatively, one can introduce an intermediate moving Σ i.e., Σ_P in which \vec{r} has a simple prescribed motion. Let \vec{r} precess round the 3-axis of Σ_P with constant angular velocity at a definite angle to it. Σ_R and Σ_P are introduced as a method of getting the solution for \vec{r} .

We shall denote the components of \vec{r} in Σ_R by single primes and in Σ_P by double primes and define Σ_R and Σ_P precisely as follows

The transformation to Σ_R is given by

$$(I \cos \phi/2 + i\sigma_3 \sin \phi/2)(r_1\sigma_1 + r_2\sigma_2 + r_3\sigma_3)(I \cos \phi/2 - i\sigma_3 \sin \phi/2) \\ = r_1'\sigma_1 + r_2'\sigma_2 + r_3'\sigma_3$$

which gives

$$r_1' = r_1 \cos \phi - r_2 \sin \phi \\ r_2' = r_2 \cos \phi + r_1 \sin \phi$$

Similarly,

$$\omega_1' = \omega_1 \cos \phi - \omega_2 \sin \phi \\ \omega_2' = \omega_2 \cos \phi + \omega_1 \sin \phi$$

Since the oscillatory field along 1-axis is a superposition of two oppositely rotating fields and since one of the components, say the clockwise, can be ignored, we have

$$\omega_1' = \frac{1}{2} |\omega_1| \cos(\omega t + \phi), \quad \omega_2' = \frac{1}{2} |\omega_1| \sin(\omega t + \phi)$$

If we prescribe for the rotating system $\phi = -\omega$... (19)

We have

$$\omega_2' = 0, \quad \omega_1' = \frac{1}{2} |\omega_1| = \frac{-\mu_{ab}}{\hbar} B_0 \quad \dots (20)$$

To find ω_3' we have from (18)

$$\dot{r}_1' = -(\omega_3 + \dot{\phi})r_2' + (\omega_1 \sin \phi + \omega_2 \cos \phi)r_3' \\ = -\omega_3'r_2' + \omega_2'r_3' \quad \dots (21)$$

Hence

$$\omega_3' = \omega_0 - \omega$$

The total angular velocity relative to Σ_R is

$$\Omega = \sqrt{\omega_1'^2 + (\omega_0 - \omega)^2} \quad \dots (22)$$

We obtain the precessional co-ordinate system Σ_P by further rotational displacements θ'' and ψ'' (about the 1-axis) and then about the new 3''-axis such that the 3''-axis coincides with the resultant angular velocity $\vec{\Omega}$ in Σ_R . We seek a transform from Σ_P to Σ_B

$$\bar{T} = \bar{T}_{\psi''} \bar{T}_{\theta''} \bar{T}_{\phi''} = \bar{T}_{\phi''} \bar{T}_{\theta''} \bar{T}_{\psi''} = T_{-\phi''} T_{-\theta''} T_{-\psi''}$$

σ_3 is then transformed to

$$\bar{T} \sigma_3 T = \gamma_1'' \sigma_1 + r_2'' \sigma_2 + r_3'' \sigma_3$$

Evaluation of the left hand side using (17) gives

$$r_1'' = \sin \theta'' \sin \psi''; r_2'' = \sin \theta'' \cos \psi''; r_3'' = \cos \theta'' \quad \dots \quad (23)$$

In fact (23) is true for a transformation to any arbitrary coordinate system, the precessional co-ordinate system in particular being defined by

$$\theta'' = \text{constant}, \quad \omega_1'' = \omega_2'' = 0, \quad \omega_3'' = \Omega \quad (24)$$

From (18) and (23)

$$\begin{aligned} \dot{r}_1'' &= r_2'' \dot{\psi}'' = \omega_2'' r_1'' - \omega_1'' r_2'' \\ \dot{r}_2'' &= -r_1'' \dot{\psi}'' = \omega_3'' r_1'' - \omega_1'' r_3'' \\ \dot{r}_3'' &= 0 = \omega_1'' r_2'' - \omega_2'' r_1'' \end{aligned}$$

which leads automatically to

$$\omega_1'' = 0, \quad \omega_2'' = 0, \quad \omega_3'' = -\dot{\psi}'', \quad \dot{\psi}'' = -\Omega t + \delta$$

where δ is a phase constant. So that

$$\begin{aligned} r_1'' &= \sin \theta'' \sin (-\Omega t + \delta) \\ r_2'' &= \sin \theta'' \cos (-\Omega t + \delta) \\ r_3'' &= \cos \theta'' \end{aligned} \quad \dots \quad (25)$$

It is easier to write explicit expressions by transforming to Σ_R for evaluation of δ . Rotating Σ_P about the 2''-axis through θ''

$$\begin{aligned} r_3' &= r_3'' \cos \theta'' + r_1'' \sin \theta'' = \cos^2 \theta'' + \sin^2 \theta'' \sin (-\Omega t + \delta) \\ r_1' &= -r_3'' \sin \theta'' + r_1'' \cos \theta'' = -\cos \theta'' \sin \theta'' \{1 - \sin (-\Omega t + \delta)\} \\ r_2' &= r_2'' = \sin \theta'' \cos (-\Omega t + \delta) \end{aligned}$$

The boundary conditions $r_1' = r_2' = 0$, $r_3' = 1$ at $t = 0$ give

$$\begin{aligned} \delta &= \pi/2 \\ \psi'' &= -\Omega t + \pi/2 \end{aligned} \quad (26)$$

and

$$\begin{aligned} r_1' &= -\cos \theta'' \sin \theta'' (1 - \cos \Omega t) \\ r_2' &= \sin \theta'' \sin \Omega t \\ r_3' &= \cos^2 \theta'' + \sin^2 \theta'' \cos \Omega t \end{aligned}$$

Again from (18) and the boundary conditions

$$\omega_2' = 0, \quad \sin \theta'' = -\omega_1'/\Omega$$

and from (22)

$$\cos \theta'' = (\omega_0 - \omega)/\Omega \quad (27)$$

Finally,

$$\begin{aligned} r_1' &= \frac{\omega_1'(\omega_0 - \omega)}{\Omega^2} (1 - \cos \Omega t) \\ r_2' &= -\frac{\omega_1'}{K} \sin \Omega t \\ r_3' &= 1 - \frac{\omega_1'^2}{\Omega^2} (1 - \cos \Omega t) \end{aligned} \quad (28)$$

This essentially solves the dynamical problem

The angles appearing in Feynman's graphical solution are just the Euler angles and are already built into the spin-1/2 formalism. The relations between the angle (denoted by θ by Feynman and Vernon)

$$\Omega t - \Omega L/v = \theta_N = \frac{L}{v} \sqrt{\omega_1'^2 + (\omega_1 - \omega)^2} \quad (29)$$

and the Euler angles ψ'', θ'' are as follows

$$\theta_N = \pi/2 - \psi'' = -\omega_1' t / \sin \theta''$$

Under conditions of exact tuning $\omega = \omega_0$ and

$$r_1' = 0, \quad r_2' = -\sin \Omega t, \quad r_3' = \cos \Omega t$$

and

$$\Omega = \omega_1', \quad \Omega t = \omega_1' t = \theta_N = \hbar^{-1} |\mu_{ab}| E_0 t \quad \dots \quad (30)$$

θ_N is proportional to the pulse strength $E_0 t$ under exact tuning

ELECTROMAGNETIC PART

Dispersion and absorption (or emission) are obtained by treating polarization P as a complex quantity (for $\Delta m = 0$ transition)

$$P_e = N(r_1' + ir_2')\mu_{ab}$$

(N , the density of molecules in the cavity) in the Maxwell equation for \vec{E}

$$\nabla^2 \vec{E} - \frac{4\pi\sigma}{c^2} \frac{\partial \vec{E}}{\partial t} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \vec{P}}{\partial t^2}$$

(σ being the conductivity). This leads to the two equations in the stationary case

$$\begin{aligned} -\omega^2 \left[\epsilon + \frac{4\pi N' \mu_{ab}}{v} \int_0^L f V^{-1} r_1' dz \right] + \omega_0^2 \epsilon &= 0 \\ -\frac{4\pi N' \mu_{ab}}{v} \omega^2 \int_0^L f V^{-1} r_2' dz + \frac{\omega_e \omega}{\Omega} \epsilon &= 0 \end{aligned}$$

where the field \vec{E} in the cavity is given by

$$\vec{E} = \vec{E}_c e(t) e^{i\omega t},$$

\vec{E}_c being the amplitude of the normal mode of the cavity, $\theta = \omega_c/4\pi\sigma$, ω_c the frequency of the cavity mode, V the volume of the cavity, f the filling factor, N' the flux of molecules travelling with velocity v

Denoting
$$A = \frac{4\pi N' \mu_{ab}}{v}$$

$$X = \int_0^L f V^{-1} r_1' dz = f V^{-1} \frac{\omega_1'(\omega_0 - \omega)}{\Omega^2} L \left(1 - \frac{\sin \theta_N}{\theta_N} \right)$$

$$Y = \int_0^L f V^{-1} r_2' dz = f V^{-1} \frac{\omega_1}{\Omega^2} v (\cos \theta_N - 1)$$

the two equations are

$$(\omega_c^2 - \omega^2)c - \omega^2 AX = 0 \quad (32a)$$

$$- \theta - c - \omega^2 AY = 0 \quad (32b)$$

Combining these two relations

$$\frac{X}{Y} = Q \frac{\omega_c^2 - \omega^2}{\omega_0} = (\omega - \omega_0) \frac{L}{v} \frac{1 - \sin \theta_N / \theta_N}{1 - \cos \theta_N} \quad (33)$$

Since we have $\omega_c \sim \omega \sim \omega_0$ under actual conditions

$$\frac{\omega - \omega_0}{\omega_c - \omega} = \frac{2Q}{\omega_0} \frac{1}{\tau} \frac{1 - \cos \theta_N / \theta_N}{1 - \sin \theta_N / \theta_N} = a \text{ (say)}$$

where

$$\tau = L/v$$

The frequency shift is then given by

$$\omega - \omega_0 = \frac{a}{1+a} (\omega_c - \omega_0) \quad (34)$$

The electric field is given directly in terms of the oscillation frequency by the definitions

$$\frac{\Omega L}{v} = \theta_N \quad \dots \quad (35)$$

and

$$\omega_1' = \frac{-\mu_{ab}}{\hbar} E_0 = \frac{-\mu_{ab}}{\hbar} V^{-1} c$$

We obtain

$$\epsilon = \frac{\hbar V^2}{\mu_{ab}} \sqrt{\frac{\theta_N^2 v^2}{L^2} - \left(\frac{a}{1+a}\right)^2 (\omega_0 - \omega_c)^2} \quad \dots (36)$$

Under exact tuning of cavity

$$\epsilon = \frac{\hbar V^2}{\tau \mu_{ab}} \theta_N \quad \dots (37)$$

The parameter θ_N itself is determined by the second of the two Maxwell relations in terms of the flux of molecules N' :

$$\frac{N'}{N_0'} = \frac{\theta_N^2}{2(1 - \cos \theta_N)} \quad \dots (38)$$

where

$$N_0' = \frac{\hbar V v^2}{2\pi |\mu_{ab}|^2 L^2 Q f^2} \quad \dots (39)$$

The power emitted under the stationary conditions is obtained by equating the loss to gain. Hence

$$\text{Power} = \frac{\omega W V}{Q} = P$$

The energy density W is given by

$$W = \epsilon^2 / 8\pi.$$

From (37) averaging over a periodic time we get under condition of exact tuning

$$W = \frac{\hbar^2 V}{8\pi \tau^2 |\mu_{ab}|^2 f^2} \theta_N^2.$$

From (38)

$$P = \frac{1}{2} N' \hbar \omega (1 - \cos \theta_N) \quad \dots (40)$$

P can also be obtained directly from r_3' , since by definition

$$P = N' \hbar \omega b b^* = \frac{1}{2} N' \hbar \omega (1 - r_3')$$

(40) is equivalent to Shimoda's result (1957).

SUMMARY AND DISCUSSION

The gas has been considered as a system of N independent molecules so that the total polarisation is taken as N times the individual dipole moment. Briefly the physical results are given by the equations (34), (36) and (40). The explicit relations of oscillation frequency, electric field and power to molecular flux are pretty complicated but are simple when the former are expressed in terms of the only parameter θ_N depending on N' as in equation (38). Approximate cases can be evaluated.

An exact analytic solution of the two-level problem is possible by the use of the spin-1/2 formalism which is suggested at the very outset by the complex

coefficients a and b which give the Cayley-Klein parameters obeying the normalization condition.

The transformation of ψ_a by the unitary matrix (3) gives the time-dependent state. Under the influence of the radiation the vector σ_3 corresponding to the initial state of the system rotates about the origin and acquires the components r_1, r_2, r_3 given by the transformation

$$U\sigma_3\bar{U} = r_1\sigma_1 + r_2\sigma_2 + r_3\sigma_3$$

where U is unitary and isomorphic with T . The transformation U is given by

$$T \rightarrow U = \begin{pmatrix} \sqrt{\frac{1}{2}(1+r_3)} & (r_1 + ir_2)/\sqrt{2(1+r_3)} \\ -\sqrt{\frac{1}{2}(1-r_3)} & (r_1 + ir_2)/\sqrt{2(1-r_3)} \end{pmatrix}$$

Since r_1, r_2, r_3 are also the direction cosines of $\vec{\mu}$

$$r_1 = \cos \alpha, \quad r_2 = \cos \beta, \quad r_3 = \cos \gamma$$

so that

$$U = \begin{pmatrix} \cos \gamma/2 & \frac{\cos \alpha + i \cos \beta}{2 \cos \gamma/2} \\ -\sin \gamma/2 & \frac{\cos \alpha + i \cos \beta}{2 \sin \gamma/2} \end{pmatrix} \quad \dots \quad (41)$$

This is the familiar rotation of σ_3 throughout an angle γ about some axis in 1-2 plane.

Alternatively the system vector can be kept fixed and the coordinate system rotated by the transformation $S \rightarrow T$ involving the independent Euler angles. This transformation gives the structure of \vec{r} in terms of the Euler angles and suggests the solution of the dynamical problem first in a "precessional" co-ordinate system and then in the rotating system Σ_R . Thus this formalism involves the three familiar representations of the rotation group in two dimensions.

The matrix T itself effects the transformation $T\sigma_3\bar{T}$ so that the r 's are defined in terms of a, b and hence the reduced density matrix is representable as a vector in spin space. The dynamical problem is to investigate its motion in spin space.

The forces of interaction appear in the transformation matrix T through the pulse strength θ_N given by (30) at exact tuning

$$T \rightarrow S = \begin{pmatrix} \cos \theta_N/2 & -i \sin \theta_N/2 \\ -i \sin \theta_N/2 & \cos \theta_N/2 \end{pmatrix} = I \cos \theta_N/2 - i\sigma_1 \sin \theta_N/2 = e^{-i\theta_N/2 \cdot \sigma_1} \quad \dots \quad (42)$$

while the response of the system is given by the transformed vector μ' whose elements are the expectation values of the unperturbed Hamiltonian and the polarization,

$$\mu = \begin{pmatrix} \langle H_0 \rangle / \frac{1}{2} N \hbar \omega_0 & \langle P \rangle^* / N |\mu_{ab}| \\ \langle P \rangle / N |\mu_{ab}| & -\langle H_0 \rangle / \frac{1}{2} N \hbar \omega_0 \end{pmatrix} \quad \dots \quad (43)$$

where $\langle H_0 \rangle = \frac{1}{2} N \hbar \omega_0 r_3$ and the quantities have to be evaluated for exact tuning $\langle P \rangle$ becomes purely imaginary. The emitted power may be obtained straight away as a difference of energies $\langle H_0 \rangle$ at $t = 0$ and $t = t$

$$P_e = \frac{1}{2} N \hbar \omega_0 - \langle H_0 \rangle = \frac{1}{2} N \hbar \omega_0 (1 - r_3') \quad \dots \quad (44)$$

Thus the spin-1/2 formalism enables one to represent the interaction process as a rotation of σ_3 about the 1-axis through the pulse angle θ_N , represented by a matrix of interaction. $-\theta_N$ is none other than the Euler angle θ in (5) through which Σ_B is rotated relative to Σ_N .

In fact no explicit reference to rotations in spin space is necessary. The relevant T -matrix can be obtained by straightforward integration from the equation

$$i\hbar \frac{dT}{dt} = HT$$

H being given by (15). r_i 's are obtained from the transformation.

$$T\rho(0)\bar{T}$$

It is not surprising that the spin-1/2 formalism is the most appropriate for the treatment of absorption and emission processes. For

$$\sigma_1 \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad \sigma_2 \begin{pmatrix} 1 \\ 0 \end{pmatrix} = i \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

$$\sigma_1 \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}; \quad \sigma_2 \begin{pmatrix} 0 \\ 1 \end{pmatrix} = -i \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

Putting $\begin{pmatrix} 1 \\ 0 \end{pmatrix} = [+]$ and $\begin{pmatrix} 0 \\ 1 \end{pmatrix} = [-]$, we simply have

$$\sigma_1[\pm] = [\mp], \quad \sigma_2[\pm] = \pm i[\mp]$$

Identifying $[+]$ as the state of the excited atom and $[-]$ as the normal atom, we note that the Pauli operators σ_1 and σ_2 are essentially 'transition operators' changing the ground state atom into an excited atom and vice-versa and play the central role in Dicke's formalism.

The gas has been considered as a system of N independent molecules so that the total polarisation is taken as N times the individual moment. Another method of treating the gas is to take the product of the T 's (42) for individual molecules and obtain a transformation for the whole gas, in which certain macroscopic operators will appear. The gas as a system of independent molecules turns out to be a special case. This will be taken up in a subsequent paper.

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